

Carrier-Density-Controlled Anisotropic Spin Susceptibility of Two-Dimensional Hole Systems

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We have studied quantum-well-confined holes based on the Luttinger-model description for the valence band of typical semiconductor materials. Even when only the lowest quasi-two-dimensional (quasi-2D) subband is populated, the static spin susceptibility turns out to be very different from the universal isotropic Lindhard-function lineshape obtained for 2D conduction-electron systems. We discuss how the strongly anisotropic and peculiarly density-dependent spin-related response of 2D holes at long wavelengths should make it possible to switch between easy-axis and easy-plane magnetization in dilute magnetic quantum wells.

Introduction – The recently achieved ability to make bulk semiconductors magnetic [1–3] opens up exciting possibilities for controlling magnetism in unconventional ways. Nanostructuring offers appealing routes towards realizing magnetic semiconducting devices based on strain-induced anisotropies [4] or wave-function engineering in heterostructures [5, 6]. In particular, size quantization strongly affects the intrinsic spin-3/2 degree of freedom carried by valence-band states [7] and is expected to stabilize easy-axis magnetism in a two-dimensional (2D) hole system with magnetization direction perpendicular to its plane [8, 9]. Our detailed theoretical study shows that the behavior of p-type magnetic quantum wells is unexpectedly rich. Valence-band mixing turns out to sufficiently weaken easy-axis magnetism at intermediate densities, triggering a transition to an easy-plane magnet. Counterintuitively, valence-band mixing is also responsible for reentrant easy-axis magnetism occurring at even higher density. Our results are robust with respect to changes in quantum-well parameters. The density-driven magnetic transitions discussed here enable new magneto-electronic device functionalities [10, 11] based on electric-field manipulation of the magnetization in low-dimensional systems.

Background & Aim – Magnetism is introduced into intrinsically nonmagnetic semiconducting materials via doping with magnetic ions [1–3] such as Mn, Co, Fe or Gd. One way to generate an exchange interaction between any two localized magnetic moments embedded in a conductor is provided by the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism [12], which gives rise to the effective two-impurity spin Hamiltonian

$$\mathcal{H}_{\alpha\beta} = -G^2 \sum_{i,j} I_i^{(\alpha)} I_j^{(\beta)} \chi_{ij}(\mathbf{r}_\alpha; \mathbf{r}_\beta) \quad . \quad (1)$$

Here $I_i^{(\alpha)}$ denotes the i th Cartesian component of an impurity spin located at position \mathbf{r}_α , and G is the exchange constant for the contact interaction between the spin density of delocalized charge carriers with the impurity spins.

The properties of the charge carriers mediating the effective impurity-spin exchange coupling are encoded in

the static spin-susceptibility tensor [12] $\chi_{ij}(\mathbf{r}_\alpha; \mathbf{r}_\beta)$. In a homogeneous electron system with spin-rotational invariance, it has a universal isotropic form that depends only on the dimensionality of the system [13], rendering the impurity-spin interaction (1) to be of Heisenberg-model type. This case applies to the 2D electron systems realized by confining carriers from the conduction band in semiconductor heterostructures as long as inversion symmetry is not broken by the crystal lattice or due to structuring of the sample [14–17].

Here we focus on the properties of 2D *hole* systems whose charge carriers have an intrinsic spin-3/2 degree of freedom that is strongly coupled to their orbital motion even when inversion symmetry is intact [7]. We adopt the Luttinger model [18] in axial approximation [7, 19, 20], which provides a useful description of the upper-most valence band of typical semiconductors in situations where

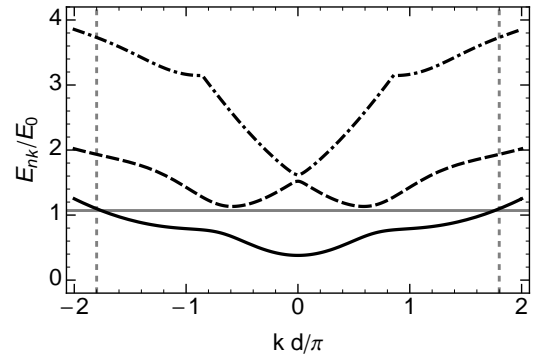


FIG. 1. Lowest three (each of them doubly degenerate) subbands of a two-dimensional hole system realized by a symmetric hard-wall quantum-well confinement of width d . Dispersions are calculated based on the four-band Luttinger-model description of bulk valence-band states in axial approximation, using band-structure parameters applicable to GaAs confined in [001] direction, and $E_0 = -\pi^2 \hbar^2 \gamma_1 / (2m_0 d^2)$. Gray lines indicate the range of energies and wave vectors for which only the lowest subband is occupied. This is the regime we focus on in this work.

its couplings to the conduction band and split-off valence band are irrelevant. Subband k-dot-p theory [21, 22] is employed to obtain the lowest quasi-2D hole subbands for a symmetric hard-wall confinement characterized by its spatial width d . See Fig. 1. We consider the case where only the lowest 2D subband is occupied and calculate the spin susceptibility. Based on this result, various magnetic phases are shown to emerge in magnetically doped hole quantum wells depending on the carrier density.

Spin susceptibility of a quasi-2D system – The spin susceptibility of 2D charge carriers is given by [13]

$$\chi_{ij}(\mathbf{R}, z; \mathbf{R}', z') = -\frac{i}{\hbar} \int_0^\infty dt e^{-\eta t} \langle [S_i(\mathbf{R}, z; t), S_j(\mathbf{R}', z'; 0)] \rangle. \quad (2)$$

Here \mathbf{R} and z are components of the position vector in the 2D (xy) plane and in the perpendicular (growth) direction, respectively. The spin density operator (in units of \hbar) is defined in terms of field operators Ψ , Ψ^\dagger and the Cartesian components \hat{J}_j of the charge carriers' intrinsic angular momentum as $S_j(\mathbf{R}, z) = \Psi^\dagger(\mathbf{R}, z) \hat{J}_j \Psi(\mathbf{R}, z)$. The field operators can be expressed in terms of operators associated with general eigenstates [labelled by band index n and in-plane wave vector $\mathbf{k} = (k_x, k_y)$] of the non-interacting Hamiltonian as $\Psi(\mathbf{R}, z) = \sum_n \int \frac{d^2k}{(2\pi)^2} e^{i\mathbf{k}\cdot\mathbf{R}} \xi_{n\mathbf{k}}(z) c_{n\mathbf{k}}$. The (normalized) spinors $\xi_{n\mathbf{k}}(z)$ and eigenvalues $E_{n\mathbf{k}}$ are obtained by solving the multi-band Schrödinger equation for the confinement in growth direction of the 2D heterostructure. We can then express the spin susceptibility as

$$\chi_{ij}(\mathbf{R}, z; \mathbf{R}', z') = \int \frac{d^2q}{(2\pi)^2} e^{i\mathbf{q}\cdot(\mathbf{R}-\mathbf{R}')} \chi_{ij}(\mathbf{q}; z, z') \quad (3a)$$

in terms of the 2D Fourier-transformed susceptibility

$$\chi_{ij}(\mathbf{q}; z, z') = \sum_{n,l} \int \frac{d^2k}{(2\pi)^2} \mathcal{W}_{ij}^{nl}(\mathbf{k}, \mathbf{q}; z, z') \times \frac{n_F(E_{l\mathbf{k}+\mathbf{q}}) - n_F(E_{n\mathbf{k}})}{E_{l\mathbf{k}+\mathbf{q}} - E_{n\mathbf{k}} - i\hbar\eta}, \quad (3b)$$

where n_F denotes the Fermi function, and

$$\mathcal{W}_{ij}^{nl}(\mathbf{k}, \mathbf{q}; z, z') = \left[\xi_{n\mathbf{k}}(z) \right]^\dagger \cdot \left[\hat{J}_i \xi_{l\mathbf{k}+\mathbf{q}}(z) \right] \times \left[\xi_{l\mathbf{k}+\mathbf{q}}(z') \right]^\dagger \cdot \left[\hat{J}_j \xi_{n\mathbf{k}}(z') \right]. \quad (3c)$$

Axial symmetry of the 2D system implies $E_{n\mathbf{k}} \equiv E_{nk}$ and permits the ansatz [23, 24]

$$\xi_{n\mathbf{k}}(z) = e^{-i\hat{J}_z \phi_{\mathbf{k}}} \bar{\xi}_{nk}(z), \quad (4)$$

simplifying calculation of the matrix elements \mathcal{W}_{ij}^{nl} . In the following, we consider the growth-direction-averaged spin susceptibility $\bar{\chi}_{ij}(\mathbf{q}) = \int dz \int dz' \chi_{ij}(\mathbf{q}; z, z')$ calculated at zero temperature.

Luttinger-model description of quasi-2D holes – Using the 4×4 Luttinger Hamiltonian in axial approximation for the bulk valence band, the bound states of 2D holes confined by a potential $V(z)$ are given in terms of spinor wave functions $\bar{\xi}_{nk}(z)$ that satisfy the Schrödinger equation $[\mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_2] \bar{\xi}_{nk} = E_{nk} \bar{\xi}_{nk}$, where

$$\mathcal{H}_0 = \frac{\hbar^2}{2m_0} \left[\gamma_1 \mathbb{1} - 2\tilde{\gamma}_1 \left(\hat{J}_z^2 - \frac{5}{4} \mathbb{1} \right) \right] \frac{d^2}{dz^2} + V(z), \quad (5a)$$

$$\mathcal{H}_1 = \frac{\hbar^2 k}{m_0} \sqrt{2} \tilde{\gamma}_2 (-i) \left(\{ \hat{J}_z, \hat{J}_- \} + \{ \hat{J}_z, \hat{J}_+ \} \right) \frac{d}{dz}, \quad (5b)$$

$$\mathcal{H}_2 = -\frac{\hbar^2 k^2}{2m_0} \left[\gamma_1 \mathbb{1} + \tilde{\gamma}_1 \left(\hat{J}_z^2 - \frac{5}{4} \mathbb{1} \right) - \tilde{\gamma}_3 \left(\hat{J}_+^2 + \hat{J}_-^2 \right) \right]. \quad (5c)$$

Here m_0 is the electron mass in vacuum, hole energies are counted as negative from the bulk valence-band edge, and we used the abbreviations $\hat{J}_\pm = (\hat{J}_x \pm i\hat{J}_y)/\sqrt{2}$, $\{A, B\} = (AB + BA)/2$. The constants γ_1 and $\tilde{\gamma}_j$ are materials-related band-structure parameters [25] and depend on the quantum-well growth direction.

Straightforward application of the subband k-dot-p method [21, 22] yields the energy dispersions E_{nk} with associated eigenspinors $\bar{\xi}_{nk}$. At $k = 0$, the eigenspinors are also eigenstates of \hat{J}_z with eigenvalues $\pm 3/2$ (heavy holes) or $\pm 1/2$ (light holes), which are split in energy. This phenomenon is often referred to as *HH-LH splitting* [7, 19]. At finite k , the spinors $\bar{\xi}_{nk}$ are not eigenstates of \hat{J}_z anymore. This phenomenon of *HH-LH mixing* arises because a spin-3/2 degree of freedom has a much richer structure than the more familiar spin-1/2 case [26]. Figure 1 shows the 2D-subband dispersions obtained for a symmetric hard-wall confinement of width d , using band-structure parameters for GaAs confined in [001] direction. The numerically obtained E_{nk} and $\bar{\xi}_{nk}$ serve as input for the calculation of the 2D-hole spin susceptibility according to Eqs. (3) with Eq. (4).

Results for the 2D-hole spin susceptibility – Using polar coordinates $(q, \phi_{\mathbf{q}})$ for the 2D wave vector \mathbf{q} and introducing the scale $\chi_0 = 2m_0/(\gamma_1 \hbar^2)$, we find that the tensor elements of $\bar{\chi}_{ij}(\mathbf{q})$ have the generic form

$$\bar{\chi}_{xx}(\mathbf{q}) = \chi_0 [F_{\parallel}(q) + G(q) \cos(2\phi_{\mathbf{q}})], \quad (6a)$$

$$\bar{\chi}_{yy}(\mathbf{q}) = \chi_0 [F_{\parallel}(q) - G(q) \cos(2\phi_{\mathbf{q}})], \quad (6b)$$

$$\bar{\chi}_{xy}(\mathbf{q}) = \chi_0 G(q) \sin(2\phi_{\mathbf{q}}), \quad (6c)$$

$$\bar{\chi}_{zz}(\mathbf{q}) = \chi_0 F_{\perp}(q). \quad (6d)$$

The functions $F_{\parallel, \perp}(x)$ and $G(x)$ depend on materials and morphological parameters of the 2D hole system, especially the hole sheet density $n_{2D} \equiv k_F^2/(2\pi)$, but $G(0) = 0$ generally. Figure 2 shows typical results obtained at low, intermediate and high densities where still only states in the lowest quasi-2D subband are occupied.

The density dependence of the 2D-hole spin susceptibility follows a generic trend. In the limit of low density [Fig. 2(a)], $\bar{\chi}_{zz}(\mathbf{q}) \gg \bar{\chi}_{xx}(\mathbf{q}) \sim \bar{\chi}_{yy}(\mathbf{q})$, and the line

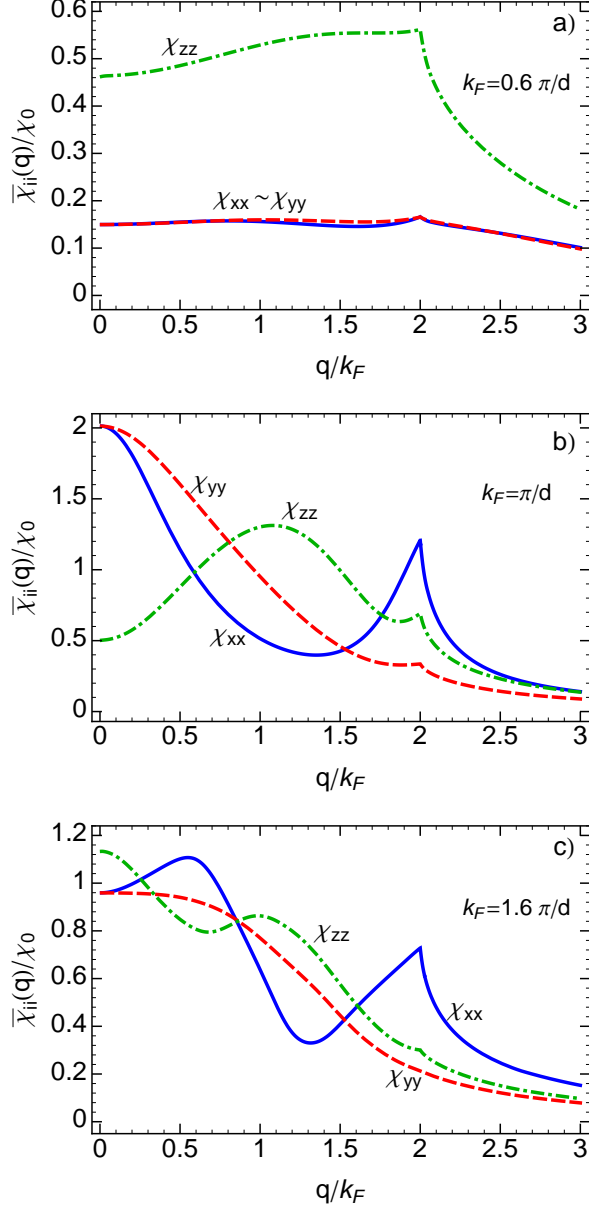


FIG. 2. \mathbf{q} -dependent spin susceptibility for $\phi_{\mathbf{q}} = 0$ and values of the 2D-hole Fermi wave vector k_F as indicated in the individual panels. Calculations are based on the band structure shown in Fig. 1.

shape of $\bar{\chi}_{zz}(\mathbf{q})$ is similar to the universal 2D-electron (Lindhard) result [13]. The strong easy-axis response is expected [6, 8, 9] as a result of HH-LH splitting, which favors a spin-3/2 quantization axis perpendicular to the 2D plane [7]. Interestingly, the behavior of the spin susceptibility changes as density is increased. For intermediate values of hole density, results like that shown in Fig. 2(b) are obtained, exhibiting *easy-plane* anisotropy in the long-wave-length limit and a nontrivial structure developing at wave vectors comparable to k_F . The significant deviation from both the universal-2D-electron

behavior and also the easy-axis response expected from HH-LH *splitting* arises because, as the 2D-hole density is increased, higher- k states get occupied that are more strongly influenced by HH-LH *mixing*. At the highest values of density where still only the lowest quasi-2D hole subband is populated, easy-axis anisotropy is restored in the long-wave-length limit but the response at finite q becomes as important in strength as that for $q \rightarrow 0$.

2D-hole-mediated magnetism – In a magnetically doped p-type quantum well, holes can mediate a magnetic interaction between localized moments via the RKKY mechanism. The corresponding spin Hamiltonian is given by Eq. (1), with $\chi_{ij}(\mathbf{r}_\alpha; \mathbf{r}_\beta) \equiv \chi_{ij}(\mathbf{R}_\alpha, z_\alpha; \mathbf{R}_\beta, z_\beta)$ being the spin susceptibility of the 2D hole system calculated here. For a random but on average homogeneous distribution of magnetic ions, standard mean-field theory [12] yields the Curie temperatures

$$T_{C_j}^{(\text{MF})} = 8\pi T_0 \left| \frac{\bar{\chi}_{jj}(\mathbf{q})}{\chi_0} \right|_{\mathbf{q}=0} \quad (7a)$$

for ferromagnetic order with magnetization direction parallel to the j axis. The temperature scale

$$T_0 = \frac{I(I+1)}{12} \frac{G^2}{k_B} \frac{n_I}{d} \frac{m_0}{\pi \hbar^2 \gamma_1} \quad (7b)$$

depends on the impurity-spin magnitude I and the average 3D density n_I of magnetic impurities, and its functional form is that obtained for 2D charge carriers with parabolic subband dispersion [9, 27] corresponding to an effective mass m_0/γ_1 .

Figure 3 shows the mean-field Curie temperatures for perpendicular-to-plane (\perp) and in-plane (\parallel) magnetization directions calculated with the same input parameters used for obtaining the subbands given in Fig. 1.

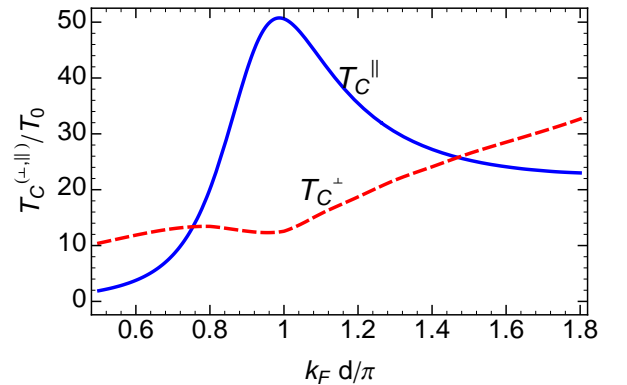


FIG. 3. Mean-field Curie temperatures for 2D-hole-mediated easy-axis (T_C^\perp) and easy-plane (T_C^\parallel) magnetism as a function of the 2D-hole Fermi wave vector k_F , obtained for a hard-wall confinement with width d and band-structure parameters applicable to GaAs.

The density dependence of $\bar{\chi}_{xx,zz}(0)$ is directly reflected in that of $T_C^{\parallel,\perp}$. At the mean-field level, the ordered state associated with the maximum transition temperature will be established. Our results suggest that the type of magnetic ordering can be modified by changing the 2D-hole density, e.g., by adjusting the gate voltage in accumulation-layer devices [28]. Easy-axis magnetism prevails at low and high densities, whereas an unexpected easy-plane magnetic order emerges at intermediate values of the density. Also for high densities, a local maximum appears in $\bar{\chi}_{xx}(\mathbf{q})$ at $q \approx 0.6k_F$, which almost reaches the value of $\bar{\chi}_{zz}(q=0)$. See Fig. 2(c). Even after averaging over the polar angle, we find that for $k_F \sim 1.5 \pi/d$ the in-plane susceptibility can have a maximum at $q \neq 0$ which is as large as $\bar{\chi}_{zz}(q=0)$. Thus it may be possible that the high-density easy-axis ferromagnetic state must coexist (or compete) with helical magnetism [12].

Finite-temperature effects – Thermal excitation of spin waves (magnons) suppresses the magnitude of the magnetization below its mean-field value M_0 . This effect is captured by the relation [29, 30]

$$\frac{M(T)}{M_0} = 1 - \frac{1}{4\pi^2 n_I d} \int d^2q \, n_{\mathbf{q}}(T) \quad , \quad (8)$$

where $n_{\mathbf{q}}(T)$ is the occupation-number distribution function of magnon modes at temperature T . A spin-wave-related critical temperature is defined by the condition $M(T_C^{(SW)}) = 0$ because, for $T > T_C^{(SW)}$, too many magnon excitations will have been excited to sustain a finite magnetization. In equilibrium, $n_{\mathbf{q}}(T)$ is given by the Bose-Einstein distribution function $n_B(\varepsilon_{\mathbf{q}}) = 1/(e^{\varepsilon_{\mathbf{q}}/[k_B T]} - 1)$, which depends on the spin-wave energy dispersion $\varepsilon_{\mathbf{q}}$. The latter's expression in terms of the charge carriers' \mathbf{q} -dependent spin susceptibility depends on the type of magnetic order (Heisenberg, Ising, or helical) [29], but the parameterisation

$$\varepsilon_{\mathbf{q}} = I G^2 \frac{n_I}{d} \chi_0 \left[\bar{\varepsilon}_0 + \bar{c}_\nu \left(\frac{q}{k_F} \right)^\nu \right] \quad (9)$$

generally holds for the relevant energy range. The dimensionless quantities $\bar{\varepsilon}_0$, ν , \bar{c}_ν depend on the functional form of the \mathbf{q} -dependent spin susceptibility and can essentially be read off Fig. 2. Stability of the magnetic order requires both coefficients $\bar{\varepsilon}_0$ and \bar{c}_ν to be positive. Inserting Eq. (9) into Eq. (8) we obtain an implicit equation for the critical temperature. Specializing to our situation of 2D-hole-mediated magnetism, we see from Fig. 2(a) that the easy-axis magnetism expected at low hole-sheet densities is actually destabilized by magnons because $\varepsilon_{\mathbf{q}} < 0$. Considering the easy-plane magnetism at intermediate densities, Fig. 2(b) reveals that the associated magnon dispersion is characterized by $\nu = 2$ and $\bar{\varepsilon}_0 = 0$, which again implies destabilization of this magnetic order due to spin-wave excitations. For the easy-axis (Ising) magnet expected at high densities [cf.

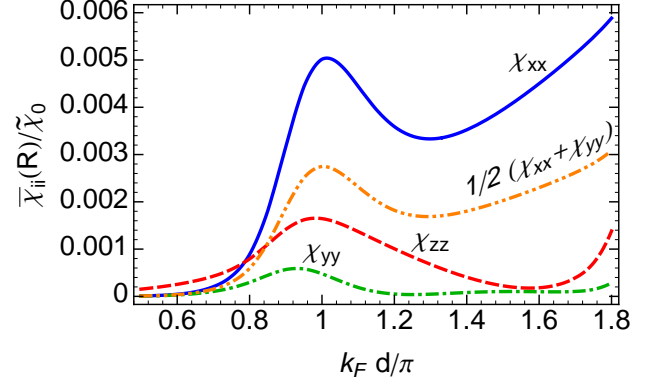


FIG. 4. Real-space spin-susceptibility $\bar{\chi}_{ij}(\mathbf{R})$ of a 2D hole system, plotted in units of $\tilde{\chi}_0 = 2\pi^2 m_0 / (\gamma_1 \hbar^2 d^2)$ as a function of $k_F d$ for fixed $\phi_{\mathbf{R}} = 0$ and $k_F R = 10$.

Fig. 2(c)], we find $\nu = 2$ and $\bar{\varepsilon}_0 > 0$. In this case a finite spin-wave-related critical temperature is obtained. Our discussion of finite-temperature effects was based on the spin susceptibility obtained for a non-interacting 2D hole system in an inversion-symmetric quantum well. However, recent studies [30] of magnetism mediated by 2D conduction-band carriers showed that the interplay of structural inversion asymmetry and Coulomb interactions can dramatically increase the stability of ferromagnetic order. As the same mechanisms exist for 2D holes, more detailed investigation of such effects will be needed to conclusively establish the stable types of confined-hole-mediated magnetic order.

Real-space spin response – Fourier transforming Eq. (6) yields the spin susceptibility in real space $\bar{\chi}_{ij}(\mathbf{R})$, which is relevant, e.g., for the proposed use of RKKY interactions to couple quantum-dot-confined spins as part of quantum-information-processing protocols [14, 16, 17]. Figure 4 shows the density dependence of $\bar{\chi}_{ij}(\mathbf{R})$ for a 2D hole system. In the low-density limit, we find $\bar{\chi}_{zz} \gg \bar{\chi}_{xx} \sim \bar{\chi}_{yy} \sim 0$, reflecting the strong easy-axis anisotropy arising due to HH-LH splitting [8, 9]. As the density of 2D holes in the lowest subband increases, in-plane components are observed to become relevant. For $k_F \sim 1.6\pi/d$ we find $\bar{\chi}_{xx}$ to be the dominant component of the spin susceptibility tensor while $\bar{\chi}_{zz} \sim \bar{\chi}_{yy} \sim 0$. Switching of the effective Hamiltonian that describes the coupling of two localized spins at a fixed distance is therefore possible by simply adjusting the density of the 2D hole system in which they are embedded.

Conclusions – Our work illustrates the crucial importance of valence-band mixing for the spin-related response in confined hole systems, which turns out to be much more strongly affected than the density response [31–33]. It also clarifies the impact of band-structure effects on hole-mediated magnetism in 2D systems which had previously been only considered for the

3D case [34–36] or outside the RKKY limit [37].

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